Coulomb Blockade and Bloch Oscillations in Superconducting Ti Nanowires

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Quantum fluctuations in quasi-one-dimensional superconducting channels leading to spontaneous changes of the phase of the order parameter by 2π, alternatively called quantum phase slips (QPS), manifest themselves as the finite resistance well below the critical temperature of thin superconducting nanowires and the suppression of persistent currents in tiny superconducting nanorings. Here we report the experimental evidence that in a current-biased superconducting nanowire the same QPS process is responsible for the insulating state—the Coulomb blockade. When exposed to rf radiation, the internal Bloch oscillations can be synchronized with the external rf drive leading to formation of quantized current steps on the I-V characteristic. The effects originate from the fundamental quantum duality of a Josephson junction and a superconducting nanowire governed by QPS—the QPS junction.

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Since the early years of superconductivity, zero resistivity and perfect diamagnetism were considered as the mandatory attributes of a superconducting state. Later it became clear that in sufficiently small systems thermodynamic fluctuations of the order parameter may significantly broaden the superconducting phase transition. In the particular case of quasi-one-dimensional (1D) superconductors [1], quantum fluctuations, also called *quantum phase slips* (QPS), enable finite resistivity in nanowires [2–8] and suppress persistent currents in tiny nanorings [9,10] at temperatures well below the critical point. Quite recently it has been realized that a superconducting nanowire governed by quantum fluctuations is dual to a Josephson junction (JJ): the Hamiltonians describing the two systems are identical with respect to parametric substitution $E_{\mathrm{QPS}} \leftrightarrow E_J$, $E_L \leftrightarrow E_C$, $I \leftrightarrow V/R_Q$, and $q \leftrightarrow \varphi$ [11]. Hence, the extensively developed physics, describing the behavior of a JJ, should be straightforwardly applicable to such a nanowire—the quantum phase slip junction (QPSJ). In the particular case of a current-biased QPSJ, it should exhibit the coherent charge oscillations qualitatively described by expressions similar to those of Bloch electrons in periodic potential of a crystal lattice. The experimental test of this prediction is the main objective of the Letter.

A conventional Josephson effect is observed in systems with coupling energy $E_J = (R_Q/R_N)(\Delta/2) \gg E_C$ and conductance $G \gg 1/R_Q$, where $R_Q = 6.45\ \mathrm{k}\Omega$, R_N is the junction normal state resistance, and Δ is the superconducting gap. In this limit the superconducting phase φ behaves as a classical variable. Application of external rf radiation with frequency f_{rf} leads to formation of quantized voltage steps on the I-V characteristic Shapiro effect: $V_n = h(f_{\mathrm{rf}}/2e)n$, $n = 0, 1, 2, \ldots$. In the opposite limit, $E_J \ll E_C$ and $G \ll 1/R_Q$, the quasicharge q rather than φ is the classical quantity, and the Coulomb effects take over the Josephson coupling [12,13]. The experimental observation of the charge phenomena in JJs requires two conditions. First, the JJ capacitance C should be small providing high charging energy $E_C = (2e)^2/2C \gg E_J$. Second, to enable the quasicharge q to be a well-defined quantity, the system should be current biased. The periodic charging of the junction leads to Bloch-type oscillations manifesting as peculiar backbended I-V characteristics. External rf radiation can be synchronized with the internal charge oscillations leading to singularities at quantized values of current $I_n = 2ef_{\mathrm{rf}}n$. Thus far, only rather broad

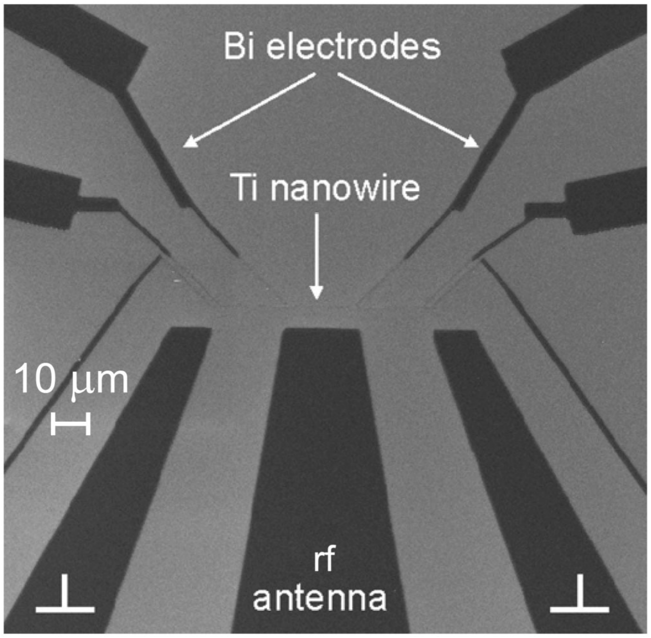

FIG. 1. Scanning electron microscope image of a typical sample with high-Ohmic contacts enabling four-probe transport measurements and introduction of rf radiation.

$n = 1$ singularities have been reported in ultrasmall JJs [14]. Indirectly, the presence of Bloch oscillations has been demonstrated in another Josephson-device–Cooper-pair box, where the injection of current I through the capacitively coupled gate resulted in formation of narrow sidebands $f_B = I/2e$ in the spectrum of the reflected rf signal [15].

Observation of the dual effect—the charge phenomena in a QPSJ—requires a sufficiently high rate of quantum fluctuations E_{QPS} exceeding the energy $E_L = \Phi_0^2/2L$ associated with the system inductance L. Here the superconducting flux quantum $\Phi_0 = 2.07 \times 10^{-15}$ Wb, $E_{\mathrm{QPS}} = \Delta(R_Q/R_N)(L/\xi)^2 \exp(-S_{\mathrm{QPS}})$, $S_{\mathrm{QPS}} = A(R_Q/R_N)(L/\xi)$, the numerical factor $A \simeq 1$, R_N is the normal state resistance of the wire with length L, and ξ is the superconducting coherence length [16,17]. If $S_{\mathrm{QPS}} \gg 1$, the rate of QPSs is small and the nanowire exhibits almost conventional superconducting properties: vanishingly small resistance below the critical current I_c. In the opposite limit $S_{\mathrm{QPS}} \simeq 1$, the quantum fluctuations are strong and, being current biased, such a (superconducting) nanowire below the certain critical voltage δV_{CB} should demonstrate the insulating state—the Coulomb blockade. Note that the model [16,17] describes the impact of rather weak quantum fluctuations, i.e., $S_{\mathrm{QPS}} \gg 1$, and has been proven to be in good quantitative agreement with experiments [7,8]. In the opposite limit $S_{\mathrm{QPS}} \simeq 1$, which is of primary interest for this Letter, strictly speaking, utilization of the model [16,17] requires further justification.

The presence of the Coulomb blockade in NbSi nanowires has been reported [18,19]. However, in this extremely high-Ohmic and strongly disordered superconductor, the presence of weak links forming a chain of JJs cannot be ruled out completely. In our work we study titanium nanostructures, where it has already been demonstrated that below the effective diameter $\sigma^{1/2} \simeq 50$ nm the rate of QPSs is sufficiently high to broaden the $R(T)$ phase transition in nanowires (with low-Ohmic environment) [8] and to suppress the persistent currents in nanorings [9]. The samples were fabricated using the same technique described in our earlier papers [7,8,20,21]. High-Ohmic probes were fabricated either from slowly evaporated at high angle titanium, showing no traces of superconductivity down to 20 mK, or from bismuth (Fig. 1) with resistance up to $R_p \sim 50$ MΩ enabling reliable current biasing of the titanium QPSJ. The extensive scanning and transmission electron, atomic force microscopic and elemental time-of-flight elastic recoil detection analyses [8,9] revealed rather conventional polycrystalline structures of the samples without obvious structural defects and with the surface roughness ±2 nm. The presence of an extended network of weak links, blocking the metal-to-metal electric current, looks rather unlikely.

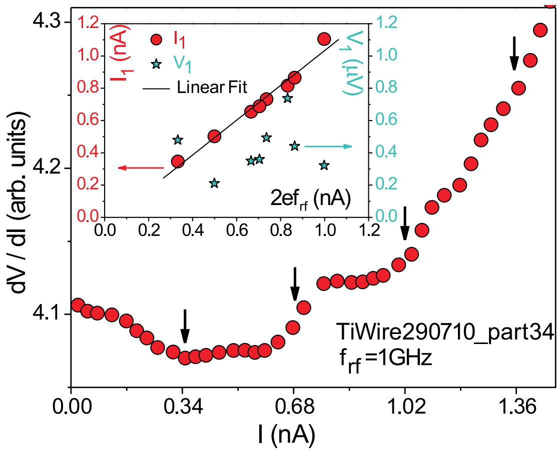

FIG. 2 (color online). All-titanium structure: The nanowire length $L = 20$ μm, $\sigma^{1/2} = 40 \pm 2$ nm, and $R_p \simeq 15$ kΩ. $dV/dI(I)$ in the presence of external rf radiation with frequency $f_{\mathrm{rf}} = 1$ GHz at $T = 70$ mK. Arrows indicate the positions of the expected current singularities $I_n = 2ef_{\mathrm{rf}}n$. Inset: Positions of the first current singularity I_1 (left axis, circles) and the corresponding voltage V_1 (right axis, stars) as function of the rf frequency f_{rf}. Note the acceptable linear fit (solid line) for the current singularities, as well as the absence of any rationality for the $V_1(f_{\mathrm{rf}})$ dependency, which one might expect in the case of a conventional Shapiro effect.

At a given temperature $T < T_c$ the relatively thick samples with diameter $\sigma^{1/2} \simeq 40$ nm and low-Ohmic probes with $R_p \simeq 15$ kΩ indeed demonstrate weak Coulomb blockade with the zero-bias conductivity lower than at a finite bias, but higher than in the normal state (Fig. 2). The observation indicates that in these samples with parameter $S_{\mathrm{QPS}} \simeq 14$ and the associated energy $E_{\mathrm{QPS}} \simeq 0.1$ μeV the residual superconductivity *wins* over the charge effects poorly resolved at realistically obtainable temperatures. Application of external rf radiation stimulates weak nonlinearities on the dV/dI dependencies at currents $I_n = 2ef_{\mathrm{rf}}n$ (Fig. 2). Note that the corresponding positions in voltage scale $V_n(f_{\mathrm{rf}})$ do not form any rational Shapiro pattern (Fig. 2, inset) to be present in a conventional Josephson system. Thinner $\sigma^{1/2} \simeq 24$ nm samples with relatively high-Ohmic $R_p \simeq 10$ MΩ probes exhibit clear Coulomb blockade with the gap $\delta V_{\mathrm{CB}} \simeq 0.6$ mV [Fig. 3(a)] corresponding to the estimation $\delta V_{\mathrm{CB}} \simeq E_{\mathrm{QPS}}$ with parameters deduced from the earlier experiments on similar titanium nanowires [8]. At small bias currents $I \leq 50$ pA the V-I characteristics of all three samples demonstrate discontinuity-type switching from the current-carrying to the insulating state. Such behavior is expected due to the non-single-valued I-V dependency [12,13]. The Coulomb gap can be quasiperiodically modulated by the gate potential $\delta V_{\mathrm{CB}}(V_{\mathrm{gate}})$ with the *rf antenna* electrode used as a dc gate [Fig. 3(a), left inset]. The period of the gate modulation is in reasonable agreement with the geometry of the experiment resulting in the highest amplitude and the smallest period for the closest to the gate sample 23, and a larger period for the remote sample 1. The

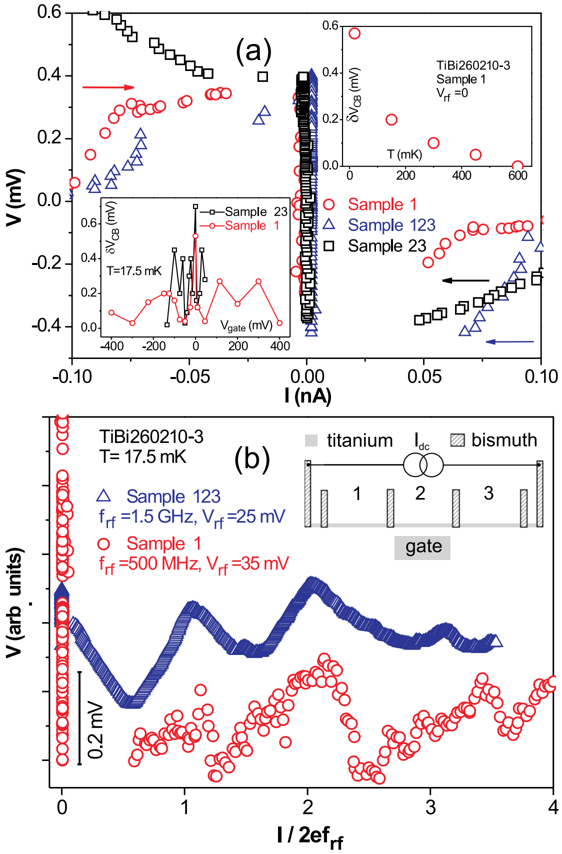

FIG. 3 (color online). Multiterminal titanium nanostructure with three adjacent nanowires each with length $L = 20\ \mu$m, $\sigma^{1/2} = 24 \pm 2$ nm, and $R_p \sim 10\ \mathrm{M}\Omega$ bismuth contacts. (a) The I-V characteristics demonstrate the Coulomb blockade for all three neighboring parts of the same nanostructure. Arrows indicate the direction of the current sweep. The Coulomb gap δV_{CB} decreases with increase of the temperature and disappears above ~450 mK (right inset). Application of the gate voltage V_{gate} quasiperiodically modulates the Coulomb gap (left inset). (b) Application of external radiation with the frequency f_{rf} generates nonmonotonic peculiarities at positions $I_n = 2ef_{\mathrm{rf}}n$. Inset: Schematics of the structure.

Coulomb gap decreases with temperature and disappears at the critical temperature of bulk titanium [Fig. 3(a), right inset]. At a given (low) temperature the Coulomb gap can be eliminated by application of sufficiently strong magnetic field. External radiation with frequency f_{rf} generates peculiarities on I-V characteristics at positions $I_n = 2ef_{\mathrm{rf}}n$ [Fig. 3(b)]. Electrodynamics of a JJ and a QPSJ is qualitatively indistinguishable. The association of our results with one of those two systems requires extra information. However, the explanation based on a conventional single electron transitor effect, due to accidental formation of tunnel junction(s) in the titanium nanowires, most likely, is not credible. First, the microscopic and elemental analyses do not reveal any presence of weak links [8,9]. Second, the effective capacitance C_{eff}, defining the charging energy $(2e)^2/(2C_{\mathrm{eff}}) = \delta V_{\mathrm{CB}}$, is about 2 orders larger than of a hypothetical parallel-plate capacitor to be formed in a break of a nanowire with ~20 nm diameter. Third, if nevertheless unintentionally the junctions were formed, it would be very unrealistic that they provide basically the same charging effect in each of the three samples: $\delta V_{\mathrm{CB}} =$ 0.6, 0.7, and 0.8 mV, respectively [Fig. 3(a)]. And fourth, the Coulomb gap disappears above the critical temperature and magnetic field for titanium. The observation does not support the interpretation based on the existence of rogue tunnel junctions, which would otherwise enable some residual Coulomb effects in the normal state. Note that the observed Coulomb gap $2e\delta V_{\mathrm{CB}}/k_B$ corresponds to a temperature of several kelvin, which is order of magnitude higher than the temperature where the last traces of the Coulomb gap disappear [Fig. 3(a), right inset]. Summarizing, in both types of structures (Figs. 2 and 3) the existence of unintentionally formed tunnel junction(s) is highly improbable, and the charge phenomena most likely originate from the QPSs providing the *dynamically driven* equivalent of a JJ—the QPSJ. The thinnest nanowires with the effective diameter $\sigma^{1/2} \lesssim 18$ nm and the parameter $S_{\mathrm{QPS}} \simeq 1$ demonstrate very pronounced back-bended I-V characteristic with the rf-induced singularities up to $n = 8$ (Fig. 4). However, the large value of the Coulomb gap [Fig. 4(a), left inset], which does not disappear above the T_c of superconducting titanium, leads to a conclusion that, though unintentionally, some weak links were formed. Note that all rf-induced singularities disappear above 200 mK, while the size-dependent critical temperature of the thinnest samples is expected to be below 250 mK [8]. At higher frequencies f_{rf} the rich structure develops at the I-V dependencies [Fig. 4(b)]. The positions of the singularities form the regular pattern [Fig. 4(c)]: $I(n,m) = e(n/m)f_{\mathrm{rf}}$, where the principal steps ($m = 1$) with $n = 1, 2, 3, 4, \ldots$ can be associated with single electron transport, while the even ones with $n = 2, 4, 6, \ldots$ can also be associated with Cooper pairs. Current singularity with $n = 1$ and $m = 2$ (indexed as "step 1/2") is resolved and formally corresponds to single electron transport. Coexistence of superconducting and single electron Bloch steps has been earlier reported in JJs, though only for $n = 1$ [22]. Remarkably, the dependence of the step width δV_n on the amplitude of the rf signal V_{rf} is essentially nonmonotonic [Fig. 4(b), inset] following the theoretical prediction $\delta V_n \sim (-1)^n J_n(V_{\mathrm{rf}})$, where J_n is the Bessel function [23]. Interpretation of the thinnest sample data (Fig. 4) is less straightforward. The nanowire sheet resistivity $R_\square$ from 0.4 to 1.9 kΩ is still on the metallic side of the metal-to-insulator transition. Coulomb effects in titanium have been observed so far in deliberately oxidized nanowires with noticeably higher resistivity [24,25]. However, the finite Coulomb gap above the T_c requires the existence of tunnel barrier(s), presumably unintentionally formed at the overlapping regions with bismuth contacts. The observed Coulomb gap [Fig. 4(a)] can be

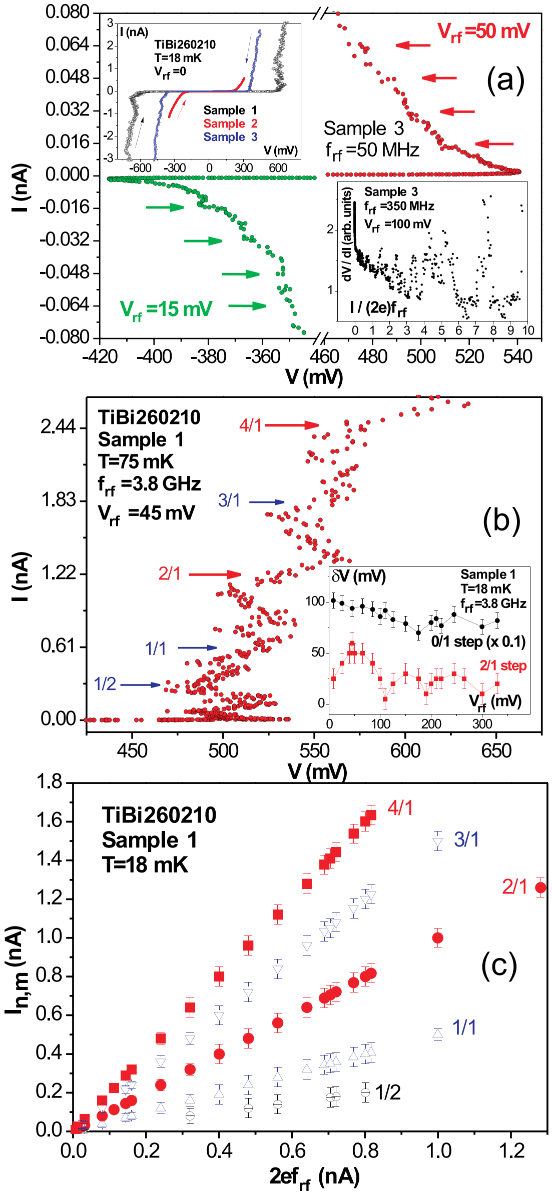

FIG. 4 (color online). Multiterminal nanostructure with each nanowire length $L = 20\ \mu\text{m}$ and the high-Ohmic bismuth contacts. (a) Sample 3 with $\sigma^{1/2} = 15 \pm 3$ nm and $R_N \approx 1.5\ \text{M}\Omega$. Zoom of the I-V characteristic at the transition point from insulating to current-carrying state at $f_{\text{rf}} = 50$ MHz and two amplitudes V_{rf} of the rf signal. Arrows indicate the positions of the expected current singularities $I_n = 2ef_{\text{rf}}n$. Note the characteristic backbended shape of the I-V dependence: the *nose*. Left inset: Larger scale I-V characteristics of the three adjacent sections of the same nanostructure. Arrows indicate the directions of the current sweep. Right inset: $dV/dI(I)$ at $f_{\text{rf}} = 350$ MHz. (b) Sample 1 with $\sigma^{1/2} = 12 \pm 4$ nm and $R_N \approx 2.5\ \text{M}\Omega$. Magnified view of the current singularities at $f_{\text{rf}} = 3.8$ GHz. Arrows indicate the positions of the expected current singularities. Inset: Dependence of the step width δV on rf magnitude for the first Cooper pair singularity (2/1) and the Coulomb gap (0/1). Note that for the Coulomb gap the scale is reduced by factor of 10. (c) Positions of the current singularities $I_{n,m}$ as functions of the rf frequency f_{rf}.

simulated by a chain ~10 parallel-plate capacitors with the area $\sigma = 15 \times 15\ \text{nm}^2$ separated by a 1 nm vacuum barrier. Note that formation of parallel junctions does not alter the Coulomb gap [26]. Hence, the major part of the 20 μm long high-Ohmic nanowires is metallic and the QPS contribution should not be disregarded. If the same number of junctions would be responsible for the Coulomb blockade and the Bloch oscillations, then the values δV_n for the steps 0/1 and 2/1 should be comparable [23], which is not the case [Fig. 4(b), inset]. One may conjecture that several serially connected junctions are responsible for the (large) Coulomb gap, while a smaller number of *active* elements is responsible for the Bloch steps. Those serially connected junctions act as an additional high-impedance environment. Given the equivalence of the quantum dynamics of a JJ and a QPSJ, our data cannot distinguish whether that active element is a static JJ or is driven by quantum fluctuations dynamic QPSJ. However, whatever is the case, our experiment is clear evidence of Bloch oscillations. It has been suggested that the proximity of a nanostructure material to a superconductor-to-insulator transition, actively studied in 2D systems [27–31], facilitates observation of the coherent QPS contribution [10]. Our present results on Ti nanowires indicate that the superconductor-to-insulator transition is not the mandatory requirement for observation of the Coulomb phenomena in QPS-driven nanowires. The resistivity of the samples is relatively low being on the metal side of the metal-to-insulator transition.

The presence of the charge effects both in thicker, essentially metallic structures (Figs. 2 and 3) and in the thinnest ones (Fig. 4), where several tunnel junctions might have been unintentionally formed, supports the universality of the phenomena originating from the fundamental duality of QPS and Josephson systems. In addition to the importance of the discovery for basic science, the observation of the Bloch singularities relating the current I_n and frequency f_{rf} through the universal relation $I_n = 2enf_{\text{rf}}$ can be considered as the proof-of-principle demonstration of the qualitatively new approach to the important metrological application—the quantum standard of electric current. Certainly, the demonstrated accuracy [Fig. 4(c)] is not yet sufficient for practical metrology. However, the high absolute values of the currents I_n are very encouraging: they reach the nA range (Figs. 2 and 4) and by far exceed the alternative single electron solutions barely providing $\sim 10^{-11}$ A currents [32]. Progress on this topic has the strong potential to revolutionalize modern metrology.

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